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Macromolecular Transport of Hydrophobic Contaminants in Aqueous Environments

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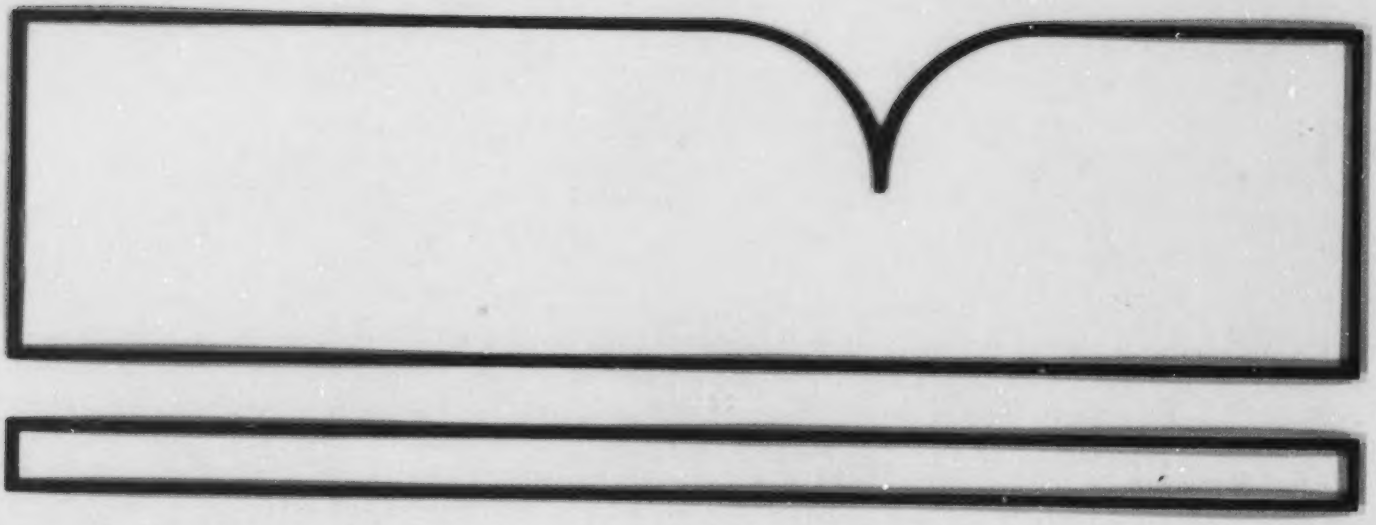

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16. ABSTRACT

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Macromolecular Transport of Hydrophobic Contaminants in Aqueous Environments

by Carl G. Enfield and Göran Bengtsson[a]

ABSTRACT

The mobility of a model macromolecule, blue dextran, was compared under laboratory conditions to the mobility of tritiated water through a sandy soil. The blue dextran eluted from the soil prior to the tritiated water. The phenomenon was compared to exclusion chromatography where molecules are separated by size with the largest eluting first and each molecule flowing through a different portion of the total porosity. The porosity occupied by the macromolecule was 0.87 times the porosity occupied by water. When the soil was amended with a mixture of kaolinite and bentonite clay in a 1:1 ratio to weight fractions of 2% and 6% total clay, the porosity occupied by the macromolecule was decreased to 0.81 and 0.66 times the porosity occupied by water, respectively. The implications to hydrophobic chemical transport based on the presence and mobility of a macromolecule were evaluated from a theoretical basis. Macromolecules should increase the relative mobility of slightly mobile compounds more than they increase the relative mobility of highly mobile compounds. Very hydrophobic compounds should show greater mobility under natural conditions than predicted, ignoring the presence of dissolved organic carbon.

INTRODUCTION

The transport of organic pollutants in a saturated porous medium is determined by many physical, chemical, and biological processes. Mathematical models recently reviewed by Rao and Jessup (1983), Boesten and Leistra (1983), and Addiscott and Wagenet (1985) describe the importance of dispersion, advection, sorption, and transformation on the movement of chemicals in soils. Fairly extensive literature (e.g., Chiou *et al.*, 1979; Karickhoff *et al.*, 1979; Kenaga, 1980; Means *et al.*, 1980; Briggs, 1981; Brown and Flagg, 1981; Karickhoff, 1981; and Schwarzenbach and Westall, 1981) has emphasized the relationship between sorption of nonpolar hydrophobic compounds, the organic carbon content of the soil, and the octanol:water partition coefficient of the sorbate. In summary, neutral organic chemicals are sorbed more strongly to soils with high organic carbon than to soils with low organic carbon. Several relationships have been developed which permit estimating a linear partition coefficient based on easily measurable properties of the chemical such as water solubility or octanol:water partition coefficient. These linear partition coefficients describe the equilibrium concentration sorbed to the organic carbon in the soil based on the concentration in the water. Multiplying the partition coefficient to the organic carbon in the soil by the weight fraction organic carbon in the soil yields the partition coefficient for the soil. Briggs (1981) proposed the relationship

$$k_d = 4.47\ F\ k_{ow}^{0.52} \qquad (1)$$

to describe the soil:water partition coefficient (k_d) as a function of the weight fraction of organic carbon in the soil (F) and the octanol:water partition coefficient (k_{ow}). Roy and Griffin (1985) suggested that relationships such as equation (1) can be expected to predict the soil partition coefficient within a factor of ten. Recently, attention has been given to the possibility of binding of the organic compounds to dissolved organic macromolecules, micelles, or other colloidal materials in the pore water. These mobile macromolecules may greatly increase chemical mobility by sorbing compounds normally sorbed by stationary soil particles.

Several studies have addressed the partitioning between an organic compound of environmental concern and dissolved organic macromolecules

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(e.g., Poirrier *et al.*, 1972; Hassett and Anderson, 1982; Carter and Suffet, 1982; Perdue, 1983; Wijayaratne and Means, 1984; Carlson *et al.*, 1985; Caron *et al.*, 1985; and Gschwend and Wu, 1985). Enfield (1985) suggested that macromolecules, or immiscible substances such as micelles or colloidal materials, moving along with a water carrier, may under certain circumstances enhance the movement of hydrophobic chemicals through soils. If hazardous chemicals partition to mobile macromolecules in subsurface environments, the current approaches of making environmental exposure and risk analysis, assuming partitioning between soil organic carbon and dilute aqueous solutions of the chemical, may significantly underestimate chemical mobility. Adverse environmental impact caused by the application of hazardous materials to the soil surface is more likely where there is a significant amount of recharge. The objective of this study was to determine the relative mobility of a model macromolecule in comparison to the mobility of water through a sandy soil and estimate, based on linear partitioning previously reported in the literature, the influence of mobility of the macromolecule on the mobility of pollutants.

THEORETICAL DEVELOPMENT

The movement of a conservative pollutant through saturated soil in the presence of macromolecules or other nonreactive organic substances can be described by dividing a representative elemental volume of the soil system into three phases: aqueous (a), solid (s), and mobile organic (o) phases. With notation given at the end of the paper, the following one-dimensional equations can be written to describe changes in mass concentration in the individual phases as a function of a dispersive flux, a convective flux, and mass transfer between adjacent phases assuming first-order kinetics.

$$\frac{\partial(\theta\rho_a C_a)}{\partial t} = D_a \frac{\partial^2(\theta\rho_a C_a)}{\partial x^2} - V_a \frac{\partial(\theta\rho_a C_a)}{\partial x} - k_{ao}\theta\rho_a C_a + k_{oa}\phi\rho_o C_o - k_{as}\theta\rho_a C_a + k_{sa}(1-n)\rho_s C_s \quad (2)$$

$$\frac{\partial[(1-n)\rho_s C_s]}{\partial t} = -k_{sa}(1-n)\rho_s C_s + k_{as}\theta\rho_a C_a \quad (3)$$

$$\frac{\partial(\phi\rho_o C_o)}{\partial t} = D_o \frac{\partial^2(\phi\rho_o C_o)}{\partial x^2} - V_o \frac{\partial(\phi\rho_o C_o)}{\partial x} - k_{oa}\phi\rho_o C_o + k_{ao}\theta\rho_a C_a \quad (4)$$

In writing the above equations, the aqueous phase was assumed to totally cover both the solid phase and the organic phase or macromolecule such that transfer of the pollutant from the organic phase to the solid phase requires passing through the aqueous phase. The total change in mass within an elemental volume can be written as the sum of equations (1) through (3) or

$$\frac{\partial(\theta\rho_a C_a)}{\partial t} + \frac{\partial(\phi\rho_o C_o)}{\partial t} + \frac{\partial[(1-n)\rho_s C_s]}{\partial t} = D_a \frac{\partial^2(\theta\rho_a C_a)}{\partial x^2} + D_o \frac{\partial^2(\phi\rho_o C_o)}{\partial x^2} - V_a \frac{\partial(\theta\rho_a C_a)}{\partial x} - V_o \frac{\partial(\phi\rho_o C_o)}{\partial x} \quad (5)$$

When local equilibrium, constant density, and constant volume fractions occupied by the different phases are assumed,

$$k_{ao}\theta\rho_a C_a = k_{oa}\phi\rho_o C_o \quad (6)$$

This yields a dimensionless organic:water partition coefficient or ratio of mass between the organic and aqueous phases as

$$k_p = \frac{k_{ao}\theta\rho_a}{k_{oa}\phi\rho_o} = \frac{C_o}{C_a}$$

Similarly,

$$k_{as}\theta\rho_a C_a = k_{sa}(1-n)\rho_s C_s \quad (7)$$

which yields a soil:water partition coefficient

$$k_d = \frac{k_{as}\theta\rho_a}{k_{sa}(1-n)\rho_s} = \frac{C_s}{C_a}$$

When field measurements are made, there is no attempt to separate the amount of chemical in the aqueous phase versus the amount associated with the macromolecules. To apply this theory to environmental situations, it is necessary to define a new variable C^* as the total mass concentration of the mobile pollutant over all phases

$$C^* = \theta\rho_a C_a + \phi\rho_o C_o \quad (8)$$

The total change in the elemental volume can be shown to follow the equation

$$\left(1 + \frac{k_{ao}}{k_{oa}} + \frac{k_{as}}{k_{sa}}\right)\frac{\partial C^*}{\partial t} = \left(D_a + \frac{k_{ao}}{k_{oa}} D_o\right)\frac{\partial^2 C^*}{\partial x^2} - \left(V_a + \frac{k_{ao}}{k_{oa}} V_o\right)\frac{\partial C^*}{\partial x} \quad (9)$$

By defining the variables

$$R^* = 1 + \frac{k_{ao}}{k_{oa}} + \frac{k_{as}}{k_{sa}} \quad (10)$$

which describes the retardation of the chemical relative to the velocity of the total fluid,

$$D^* = D_a + \frac{k_{ao}}{k_{oa}} D_o \qquad (11)$$

the effective dispersion coefficient for the total fluid, and

$$V^* = V_a + \frac{k_{ao}}{k_{oa}} V_o \qquad (12)$$

the effective velocity of the total fluid, equation (9) becomes

$$R^* \frac{\partial C^*}{\partial t} = D^* \frac{\partial^2 C^*}{\partial x^2} - V^* \frac{\partial C^*}{\partial x} \qquad (13)$$

Equation (13) is the same form that has been solved analytically for a variety of boundary and initial conditions. Several of these solutions have been summarized by van Genuchten and Alves (1982). The theoretical development presented includes retardation of the chemical due to partitioning between the phases and the influence of partitioning on the dispersive and advective transport of a hydrophobic chemical when macromolecules are present.

MATERIALS AND METHODS

The soil for the experimental portion of the study was obtained from the current surface horizon of an infiltration basin used at the Vomb water plant to treat surface water for the drinking-water supply of the cities of Lund and Malmo, southern Sweden. The infiltration basins are natural sand deposits. The system operates by intermittently applying lake water to the surface of the infiltration basin, and the filtered water is extracted using wells. The study soil sample had no visible structure in nature and was collected as a disturbed sample. The soil was air-dried and homogenized as much as possible prior to characterization or experimentation. The weight fraction total organic carbon of the soil was determined, on ten replicate samples, using a (Leco model CR-12) total carbon analyzer.

Blue dextran, a polysaccharide synthesized from sucrose by Lactobacillaceae, was selected as a model compound for the study. The size and hydrophilic characteristics of this compound are similar to natural microbial exopolymers found in soil and ground-water systems. This type of hydrophilic carbon compound is not normally identified in aqueous solutions even under rigorous GC-MS evaluations. The manufacturer, Pharmacia Fine Chemicals, reports the molecular weight of this compound to be approximately 2,000,000 g $mole^{-1}$. The octanol:water partition coefficient (k_{ow}) for blue dextran was determined by equilibrating 10 ml of ground water containing 0.05 μmol (100 mg l^{-1}) dextran with 10 ml of n-octanol for 24 hours. After equilibration, the mixture was centrifuged at 3000 g for 20 minutes. The amount of dextran in the octanol was measured, at a wave length of 630 nm, with a (Beckman DB-GD) grating spectrophotometer.

The experimental columns, made from stainless steel, were 5 cm in diameter and 3 cm in length. The top and bottom of the soil sample were supported with sintered stainless steel plates. The replicate columns were packed to a bulk density of 1.5 Mg m^{-3}. Assuming a particle density of 2.65 Mg m^{-3} for the mineral soil, the porosity (n) would be 0.43. After establishing steady-state hydraulic conditions with ground water collected from one of the adjacent extraction wells near the infiltration basin where the soil was sampled, a solution containing 0.25 μmol (500 mg l^{-1}) of dextran and 2 μCi l^{-1} tritiated water, in ground water, was pumped through the soil for a period of 3.67 hours. The Darcy flow velocity was approximately 50 cm d^{-1}, corresponding to the average infiltration rate at the water treatment facility. The pulse of dextran and ^3H was followed by ground water for several additional pore volumes. The pulse length was selected such that the effluent concentration would equal the influent concentration prior to removing the pulse. Dextran and ^3H were monitored as a function of time in the column effluent. Samples were collected in glass tubes using a Fractovap fraction collector every 8 min. The volume of each sample was approximately 5.5 ml. One ml from each fraction sampled was mixed with Beckman CP cocktail in 7-ml polyethylene vials and counted for 10 min on a (Beckman model 1801) scintillation spectrophotometer. Dextran was measured in each sample as indicated earlier.

RESULTS AND DISCUSSION

Representative experimental breakthrough curves for the tritiated water and blue dextran are shown in Figure 1. Tritiated water is commonly used in laboratory experiments to describe water movement through soil columns. The experiment was designed such that water-filled porosity could be obtained directly from the experimental data without obtaining solutions to equation (13). The volume of water in the column (one pore volume) is equal to the volume of water required to increase the effluent tritiated water concentration from zero to 0.5 times the influent concentration with both influent and effluent concentrations corrected for background decays. The approach is valid for a conservative nonretarded chemical, such as tritiated

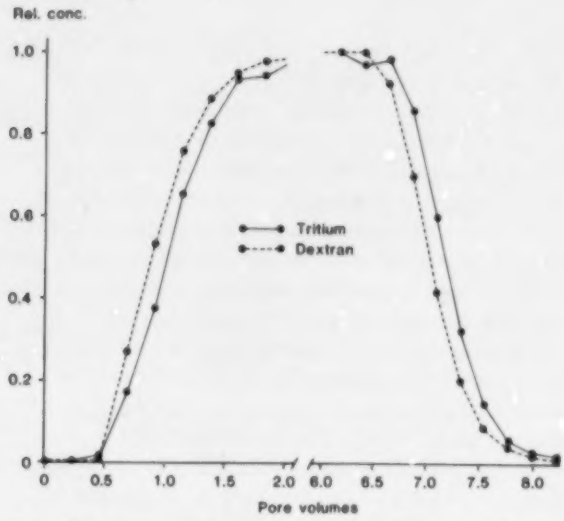

Fig. 1. Relative concentration of dextran and ^3H versus pore volumes. The pore volumes were determined gravimetrically assuming a particle density of 2.65. Corrections were made to minimize errors due to lengths of inlet and outlet tubing.

water, when the effluent concentration is allowed to reach the influent concentration prior to removing the chemical pulse. In Figure 1, the data were normalized to pore volumes and relative concentrations. One pore volume determined gravimetrically was consistent with volume required to reach a relative concentration of 0.5 for the tritiated water. The octanol:water partition coefficient for the blue dextran was experimentally determined to be 0.09. From equation (1), one can estimate, based on the octanol:water partition coefficient and the measured weight fraction of carbon in the soil (0.002 ± 0.0003 or 0.2 percent), that the soil:water partition coefficient should be approximately 0.0026. If the blue dextran is miscible with water and does not behave as a separate phase, the retardation factor from equation (10) reduces to

$$R = 1 + [(1 - n)\rho_s/n\rho_a]k_d \qquad (14)$$

The estimated retardation factor for blue dextran in the study soil becomes 1.009, and one would anticipate the blue dextran to elute from the column slightly behind the tritiated water. The blue dextran, nevertheless, eluted from the column prior to the tritiated water. The experiment was repeated three times on four different soil columns packed to the same bulk density. A relative concentration of 0.5 was observed at 0.87 ± 0.01 pore volumes as shown, for one data set, in Figure 1. Apparently the large dextran molecule was excluded from the smaller soil pores and forced to move through only the larger pores. This suggests that the effective pore volume for this macromolecule soil combination was approximately 13 percent less than for the tritiated water, and the resultant average interstitial velocity of the blue dextran would be greater than for the water. The observations are similar to those in gel chromatography (Giddings and Mallik, 1966) where chemicals are separated by size with the largest molecules being eluted first, followed by successively smaller molecules.

To further test the hypothesis that the macromolecules were being excluded from the smaller pores, subsamples of the Vomb soil were amended with commercial samples of kaolinite and bentonite clay. One sample of soil was amended such that it contained 2 volume percent clay; 1 volume percent each of kaolinite and bentonite. A second sample of soil was amended such that it contained 6 volume percent clay; 3 volume percent each of kaolinite and bentonite. The soils were homogenized, by tumbling, prior to packing. After establishing hydraulic and ionic steady-state conditions with ground water assumed after 100 pore volumes, similar tests were performed as described above except the Darcy flow velocity was reduced to 14.7 cm d^{-1}. The blue dextran obtained a relative concentration of 0.5 at 0.81 and 0.66 pore volumes, respectively, for the clay amended soils. The data collected demonstrates that large hydrophilic molecules can be excluded from smaller soil pores and thus appear to move with a higher average velocity than the water. The data presented are not sufficient to determine the size of pore that is excluded by this macromolecule or suggest that all macromolecules of the same weight would behave in a similar manner. The experimental data demonstrate a phenomenon which should be considered when performing exposure assessments.

Two theoretical figures were developed to elucidate the significance of the presence of the macromolecule and the differences in interstitial velocities between fluid phases for the transport of trace organic pollutants. Several simplifying assumptions were made in the development of the figures including: (1) dispersion in equation (13) was ignored which yields

$$R^* \frac{\partial C^*}{\partial t} = -V^* \frac{\partial C^*}{\partial x} \qquad (15)$$

(2) the density of the macromolecule equals the density of the water; (3) the partition coefficient to the macromolecule organic carbon is the same as

the partition coefficient to the soil organic carbon (k_{oc}), $k_p = k_{oc}$ times the weight fraction of carbon occupied by the macromolecules; (4) the weight fraction occupied by the organic carbon (F) is 0.02; (5) the soil partition coefficient can be described by equation (13); (6) the total porosity (n) of the soil is 0.5; and (7) the particle density of the soil is 2.65.

The importance of the macromolecule on the mobility of hydrophobic compounds is estimated in Figure 2. If the interstitial velocity of the macromolecule and the water are the same ($V_a = V_o$), the octanol:water partition coefficient versus the relative chemical mobility (mobility of hydrophobic compounds with macromolecule / mobility without macromolecule) can be presented. A group of chemicals with different k_{ow} are shown on the figure for reference purposes only. Figure 2 displays the change in mobility of hydrophobic chemicals due to the macromolecule rather than the mobility of the chemical. Macromolecules in the mobile phase can significantly alter the relative mobility of extremely hydrophobic compounds even when the amount of macromolecule is in concentrations typical of ground water or agricultural soil solution (1 to 10 mg l^{-1} dissolved organic carbon of unknown composition). This might be one explanation of why hydrophobic pesticides such as DDT have been reported to move farther under field conditions than model projections (e.g., Enfield *et al.*, 1982). The importance of the macromolecule rapidly diminishes as the octanol:water partition coefficient goes down. At municipal or industrial waste sites where relatively high concentrations of macromolecules might exist, the importance of the macromolecule to chemical mobility could be significant even for chemicals with log k_{ow} of 3 or less. Hydrophilic macromolecules similar to dextran at carbon concentrations of 100-500 mg l^{-1} may change the relative mobility of hydrophobic compounds by an order of magnitude in low carbon soils.

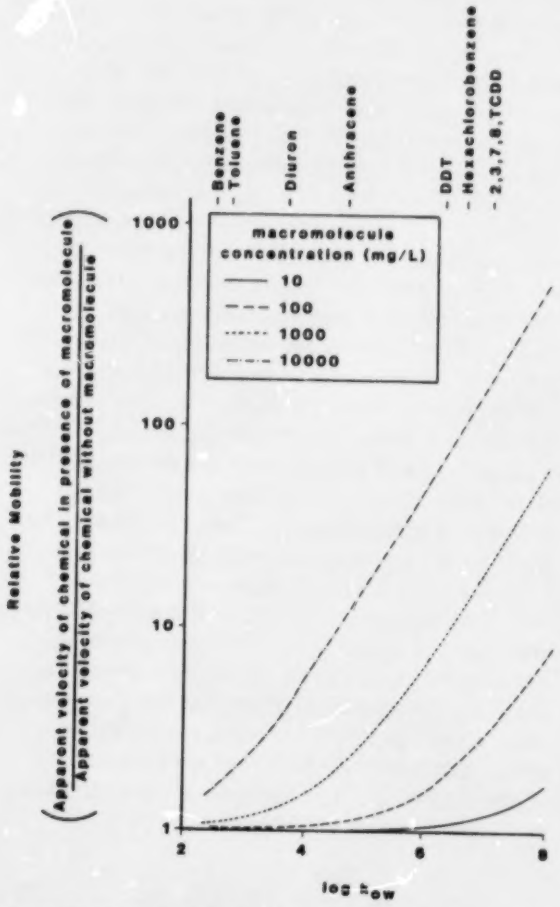

Fig. 2. Mobility of a hydrophobic compound relative to the mobility of the same compound without the presence of a macromolecule as a function of octanol-water partition coefficient and amount of organic carbon in the mobile phase.

The significance of differences in interstitial velocities of the two mobile phases are shown in Figure 3. It was assumed, in Figure 3, that the carbon concentration of the macromolecule was 100 mg l^{-1}. It is possible to plot relative mobility of the macromolecule (V_o/V_a) versus the relative mobility of the trace compound for different interstitial velocities of the mobile phases. The increased velocity of the macromolecule itself may increase the mobility of hydrophobic trace compounds by a factor of 5 or 10. It can be seen from the figure that if the velocity of the macromolecule drops to 10 percent of the water velocity, the importance of the macromolecule in accelerating transport approaches zero. Therefore, chemical transport will be facilitated only by macromolecules which behave as hydrophilic compounds (have high mobility).

The soil selected for study was a sandy soil and expected to have only a small proportion of its pores that could not be penetrated by the macromolecule. The observed decrease in effective porosity of the natural soil would not have a major impact on the mobility of chemicals at this site. However, under conditions where there is a high percentage of small pores, as would be anticipated in clay liners used in the construction of lagoons, there could be a significant increase in the mobility of chemicals due to a reduction in effective porosity

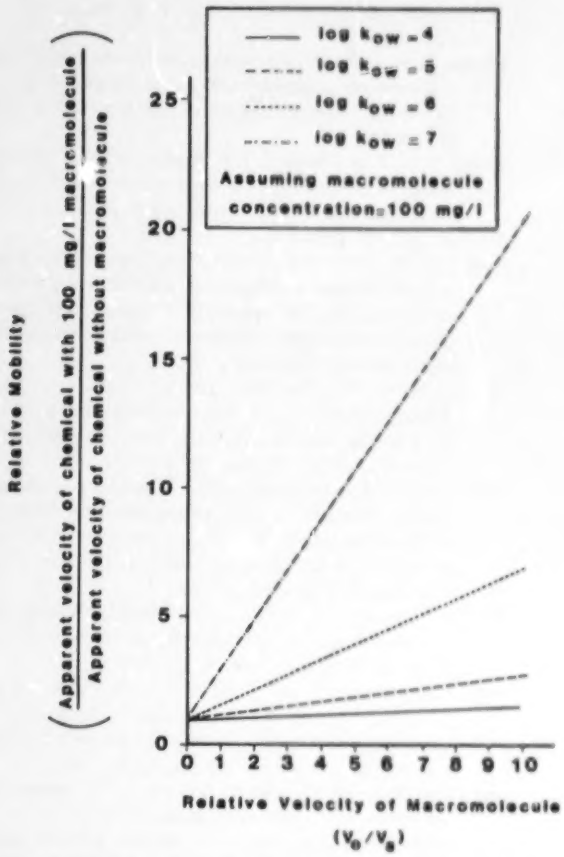

Fig. 3. Mobility of a hydrophobic compound relative to the mobility of the same compound without the presence of a macromolecule versus the velocity of the macromolecule relative to the velocity of the water where the concentration of the macromolecule is 100 mg l^{-1}.

to the macromolecule, as demonstrated by the additions of clay to the Vomb soil. This could have significant implications at industrial holding ponds and lagoons. Even when the macromolecule does not move faster than the water, it should dramatically increase the mobility of chemicals. There are two important implications to the macromolecule: (1) When a municipal landfill which produces large amounts of dissolved organic carbon is codisposed with toxic hydrophobic chemicals, the large amounts of DOC produced by the natural degradation processes will create conditions favorable to the transport of hydrophobic materials normally considered to be immobile. (2) At locations where the soils are already contaminated with hydrophobic materials, the addition of macromolecules to the system potentially will reduce the time required to remove the hydrophobic material from a contaminated aquifer.

NOTATION

C_a Mass concentration in aqueous phase, g g^{-1}.

C_o Mass concentration in immiscible phase, g g^{-1}.

C_s Mass concentration in solid phase, g g^{-1}.

C^* Mass concentration of mobile phase, g g^{-1}.

D_a Dispersion in aqueous phase, m^2 d^{-1}.

D_o Dispersion in immiscible phase, m^2 d^{-1}.

D^* Lumped dispersion coefficient, m^2 d^{-1}.

F Fraction of organic carbon associated with the soil.

k_{ao} First-order transfer coefficient aqueous to immiscible phase, d^{-1}.

k_{as} First-order transfer coefficient aqueous to solid phase, d^{-1}.

k_d Soil:water partition coefficient.

k_{oa} First-order transfer coefficient immiscible to aqueous phase, d^{-1}.

k_{oc} Organic carbon:water partition coefficient.

k_{ow} Octanol:water partition coefficient.

k_p Mobile carbon:water partition coefficient.

k_{sa} First-order transfer coefficient solid to aqueous phase, d^{-1}.

n Volume fraction of soil voids (porosity), m^3 m^{-3}.

R Retardation factor.

R^* Lumped retardation factor.

t Time, d.

V_a Interstitial velocity of aqueous phase, m d^{-1}.

V_o Interstitial velocity of immiscible phase, m d^{-1}.

V^* Lumped interstitial velocity of mobile phase, m d^{-1}.

x Distance along flow path, m.

θ Volume fraction occupied by the aqueous phase, m^3 m^{-3}.

ρ_a Density of aqueous phase, Mg m^{-3}.

ρ_o Density of immiscible phase, Mg m^{-3}.

ρ_s Particle density of the soil, Mg m^{-3}.

ϕ Volume fraction of the immiscible phase, m^3 m^{-3}.

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